

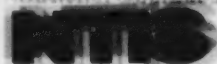
Photochemical Oxidant Formation
Overview of Current Knowledge and
Emerging Issues

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PHOTOCHEMICAL OXIDANT FORMATION:
Overview of Current Knowledge and Emerging
Issues

by

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PHOTOCHEMICAL OXIDANT FORMATION: OVERVIEW OF
CURRENT KNOWLEDGE AND EMERGING ISSUES

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ABSTRACT

Despite 1-1/2 decades of control effort, the photochemical ozone problem continues to plague human society and ecology in the US. One reason alleged for the difficulty in achieving the established ozone air quality standard is that current understanding of the science underlying the problem is still insufficient. Scrutiny of existing empirical and theoretical/experimental evidence revealed imperfections in ozone air quality models and model application procedures. Latest chemical mechanism developments established the CBM and CALL mechanisms as superior to others but still lacking in some respects. Precursor-related uncertainties are of consequence both for models requiring emission rate input and for models requiring ambient concentration input. Other factors affecting model predictions are the composition of VOC emissions and the amount and makeup of pollutants transported into the modeled atmosphere. Regional ozone air quality models are now available and procedures are being developed for use of such models in formulating regional ozone control strategies. The relative importances of "hydrocarbons" (VOC) and nitrogen oxides (NO_x) differ in the urban and the regional ozone formation phenomena.

INTRODUCTION

It has been some 3 1/2 decades since Haagen-Smit and co-workers (ref. 1) demonstrated that the smog symptoms experienced typically in Los Angeles and other urban areas are products of photochemical reactions of organic (VOC) and inorganic (NO_x) pollutants. Since then, several notable developments have taken place:

- The specific chemical species responsible for the smog effects (eye-irritation, plant/materials damage, etc.) have been identified (e.g. O_3 , PAN, aldehydes, H_2O_2 , etc.) (ref. 2).
- Organic emissions have been found to differ widely in ozone-forming potential, a finding that led to the concept of discriminate VOC control for ozone reduction (ref. 3).
- Ozone and other pollution problems have been found to be a regional rather than an urban scale phenomenon as a result of multi-day pollutant transport (ref. 4).

- The mechanism of the atmospheric chemical process that produces ozone has been elucidated in great detail; it now takes hundreds of elementary chemical reaction steps to describe the mechanism as now understood (refs. 5-7).

- Urban scale and regional scale ozone air quality models capable of predicting spatial and temporal occurrence of ozone have been developed and are now in use (ref. 8).

These remarkable advancements and the fact that they required more than 35 years of focused research effort appear to suggest that current understanding of the photochemical ozone problem is adequate and attention naturally shifts to "newer" emerging air pollution problems (e.g. toxic pollution, acid rain and problems arising from changes in stratosphere and global climate). However, the latest efforts to evaluate mechanistic models and the disappointing experiences in the US from application of O₃-related controls have all indicated that there are still serious gaps in our understanding of the ozone problem. Ozone research, and further advancement of the theory underlying the ozone problem, clearly, must continue until there is convincing evidence that theoretical predictions agree with real world observations. In the discussion that follows, an attempt will be made to summarize the state-of-science in the ozone problem area and to identify and discuss outstanding scientific issues.

STATE-OF-SCIENCE AND ISSUES

It is now accepted that the main reason for not having solved the ozone problem after 35 years of research is the enormous complexity of the problem. This complexity stems from the facts that O₃ is an extremely reactive pollutant and that it can be scavenged by the very same pollutants that produce it. It is for this latter reason that the ozone concentrations do not respond linearly to precursor controls or to dilution. Therefore, the key prerequisite to solving the ozone problem is a good, quantitative understanding of the ozone-to-precursor relationships, which, in turn, requires a good, detailed understanding of the atmospheric ozone-forming process.

Ozone to precursor relationships (ref. 2) are derived from basic laboratory, smog chamber, and modeling evidence, and to a lesser extent from empirical, field monitoring studies also. Unlike in past years, smog chamber data today are not used directly to derive such relationships. Instead, such data are used only for the purpose of developing or evaluating chemical mechanisms. For this reason, smog chamber data, along with laboratory kinetic and mechanistic data are now viewed as part of what is commonly referred to as modeling evidence, in distinction to empirical, field monitoring evidence. How do the modeling evidence and empirical evidence rate relative to each other?

Recent studies by the State of California illustrate the type and utility of empirical evidence pertaining to the ozone problem (ref. 9). Notwithstanding its non-causative nature and other limitations, such evidence is believed by many to have indisputable merit because of the intrinsic validity associated with real world data. For the evidence to be conclusive, however, there must be an abundance of data, e.g. multi-year historical data on emissions, air quality and meteorology, and the data must be of documented reliability. Based on experiences to date, these conditions only rarely can be met; for that reason the amount of acceptable empirical evidence in existence is rather limited and can only be of suggestive nature. In contrast, the modeling evidence is abundant and extremely useful for deriving quantitative ozone-to-precursor relationships. In fact, the relationships we currently accept and use have been derived entirely from modeling evidence and for that reason, the next logical question is how reliable and complete this evidence is.

In judging the reliability and completeness of the modeling evidence, specific items of interest are the accuracy of the chemical and transport/dispersion mechanisms for the atmospheric ozone formation process, the accuracy of the requisite chemical kinetic data, the accuracy of the precursor (ambient) concentration and emission inventory data, the distinction between "reactive" versus "non-reactive" VOC's, and the role of pollutant transport.

In 1981, we learned from sensitivity studies that different chemical mechanisms appearing to have equally good credentials, when used to compute VOC control requirements, could give widely different results (ref. 10). This was disturbing and underlined the need to develop more reliable methods for evaluating chemical mechanisms. Such methods--e.g. checking mechanisms against smog chamber data or field data, or against theory--do exist and, in concept can give useful evaluations provided the requisite data and theory are sufficiently accurate and comprehensive. In a 1986 workshop in USA on Evaluation of Chemical Mechanisms, the participants agreed that, despite current shortcomings of smog chamber data, testing of a chemical mechanism against such data is still the most reliable and indispensable method for evaluating mechanisms (ref. 11). Smog chamber data are not without shortcomings. The most important, well known shortcoming is that smog chambers exhibit erroneously high reactivity because of chamber wall effects. The question of how the chamber walls generate this extra reactivity has not been fully and satisfactorily answered yet. Very recently (1987), Dr. Akimoto and his co-workers at the Japanese Institute for Environmental Studies reported evidence suggesting that the extra reactivity is due to a photoenhanced surface reaction between NO_2 and H_2O which results in production of nitrous acid some 2-6 times more than originally thought (ref. 12). It would be very useful if other smog chamber researchers would react to or test this explanation so that it can be determined whether the Japanese theory has broad validity.

From efforts to date in the US, two mechanisms have emerged as having the best credentials because of their long history of development and testing. These are: SAI's Carbon Bond Mechanism (CBM) (ref. 5), and the

Carter-Atkinson-Lurmann-Lloyd (CALL) mechanism developed by U. California and ERT researchers (ref. 6). Ozone predictions by the two mechanisms agree well with each other and they also agree equally well--or equally badly--with existing smog chamber data. The two mechanisms have undisputed strengths but they also have imperfections (ref. 11). For example, the portions of the mechanisms that describe the reaction of OH with most paraffins, olefins and aldehydes have been shown to be accurate. The mechanisms, however, have uncertainties in the areas of aromatic hydrocarbon chemistry, the chemistry of heavy alkanes, and the ozoneolefin reaction chemistry. Uncertainties are also caused by the lack of photolytic data for several carbonyl compounds, e.g. MEK, propionaldehyde, acetone, glyoxal, and methylglyoxal. One piece of evidence attesting to presence of imperfections is the fact that the O₃ predictions of the 2 mechanisms respond differently to changes in VOC composition.

One factor that has been drawing increasing attention in modeling studies of the ozone problem is the precursor emissions or ambient concentrations that are input to the models. Use of existing emission inventory data in air quality models to compare model-computed ambient VOC concentrations with observations or to compare VOC-to-NO_x emission ratios to respective ambient concentration ratios or to compare anthropogenic-to-biogenic VOC emission ratios to respective ambient ratios resulted in inconsistencies that suggest erroneously low emission inventories for anthropogenic VOC's (refs. 13,14). More recent evidence on heretofore ignored anthropogenic VOC emissions (refs. 15,16) [e.g. from TSDF, POTW's, fugitive emissions, higher evaporative auto emissions, etc.] has verified this emission problem and has cast a greater doubt on the accuracy of model predictions regarding control requirements for ozone. Another emission-related problem is the lack of reliable VOC emission composition data with detail consistent with the model's requirements. Given the sensitivity of model predictions to VOC composition, this problem is not insignificant.

Problems also exist in the cases for which the main precursor-related inputs to the model are ambient concentrations. A key input to the EKMA model is the 6:00-to-9:00-am VOC-to-NO_x concentration ratio, an entity which, as presently measured--i.e. at center-city sites during 6-9 AM--is of questionable validity. To explain, such ratios vary widely from location to location (within the center-city area) and from day to day both because of measurement error and because of normal variability of the source and meteorology factors. Because of the latter reason, selecting the average or mean ratio value (as is currently done) for inputting to the EKMA model may or may not be justified. In efforts to test the validity of the current method and to search for more valid alternatives, a field study is being currently conducted by USEPA in which measurements of VOC and NO_x are made in the same air mass where the day's peak O₃ concentration occurred. Such VOC and NO_x measurements, of course, do not represent the 6-9-AM concentrations--some VOC and NO_x must have reacted out and some fresh VOC and NO_x must have mixed in during the time in which ozone reached its peak concentration. Some adjustment for these effects, however, are possible. For example, measurement of NO_x reaction products can provide a basis for

adjusting the VOC/NO_x ratio for reaction. The data from this study will be examined for evidence on the merits of this method for obtaining the requisite VOC/NO_x value.

The distinction between reactive and non-reactive VOC's has significance because only reactive VOC emissions are required to be inventoried and because reactivity must be considered in judging impacts of VOC emission trade-offs (e.g. emissions from alcohol-powered autos versus emissions from gasoline autos). Several reactivity classification schemes have been conceived to date, ranging from the simplest but least effective 2-Class (reactive-unreactive) scheme to a tedious to use but more effective 5-Class scheme (ref. 2). Current thinking in the US favors a 2-Class scheme that uses the reactivity of ethane as the "borderline" separating reactive from non-reactive VOC's. In most--but not all--cases, a VOC can be reactivity-characterized through experimental measurement of its rate of reaction with the OH radical (ref. 17). For some VOC's, however, (e.g. high molecular weight aliphatics) the OH reaction rate constant is not a valid index of ozone-forming potential (ref. 18), and use of more complex experimental/modeling techniques is required.

With respect to pollutant transport, the phenomenon has been studied to the point that regional O₃ air quality models are now available. It has been established that long range (i.e. multi-day) pollutant transport influences the O₃-to-precursor relationships drastically. Under regional or multi-day transport conditions, for example, increasing NO_x consistently enhances O₃ formation (ref. 19) whereas under single day transport conditions increasing NO_x may either enhance or inhibit O₃ formation (depending on VOC/NO_x and other conditions) (refs. 20,21).

Regional O₃ models have been developed (ref. 8) but have not been fully evaluated yet. In fact, the modelers have not devised yet a totally satisfactory method for evaluating regional models. Comparing regional model predictions and observations in pairs, as commonly done with urban scale models, is much less meaningful because of the more strongly stochastic nature of the regional model predictions. Statistical comparison of predictions and observations (ref. 22), although somewhat less useful, is more rational and appears to be the preferred evaluation approach at this time.

While most of the ozone in problem areas originates from photochemistry of urban emissions, significant contributions can arise also from stratosphere-troposphere exchanges, from background tropospheric chemistry, from biogenic emissions, and from photochemistry of diffuse anthropogenic emissions in rural areas (ref. 23). The sum total of these contributions, referred to as "background ozone", can reach significant levels in certain areas and for that reason it warrants attention. Tropospheric chemistry has been reported to result in ozone formation only when tropospheric NO_x concentrations exceed a certain threshold value; otherwise, destruction of ozone occurs (ref. 24). Biogenic VOC's have been studied and found to be both potent producers and extremely effective scavengers of ozone (ref.

24). There have been studies suggesting that biogenic VOC's have an insignificant role in urban ozone problems (ref. 24). They may contribute, however, to rural ozone--an effect which is supported by some but not enough evidence (ref. 25). Finally, existing evidence on the significance of rural anthropogenic emissions as a source of background ozone is limited and somewhat unclear. Modeling estimates vary depending on NO_x assumptions and/or calculation techniques used (ref. 25). Thus, the questions of magnitude, origins and controllability of background ozone are open and important and call for additional research.

Next, the discussion will address the control implications of the recent advances in O_3 chemistry and modeling, and some control-related questions that remain unanswered due to unresolved scientific issues.

Recent modeling studies have established that the relative roles of VOC and NO_x in urban O_3 formation vary considerably from city to city depending on a host of factors the most important of which is the VOC-to- NO_x ratio (ref. 20). The relative merits of VOC control and NO_x control depend also on whether the control goal is an immediate and modest reduction of peak O_3 concentrations or the drastic O_3 reduction required to meet the O_3 air quality standard. For example, NO_x control can bring about an immediate modest reduction of peak O_3 concentration in urban areas but it may also, under some, not uncommon conditions, make it more difficult to ultimately achieve the O_3 air quality standard (ref. 20). Finally, the evidence indicates that whereas VOC control is never detrimental, NO_x control can be detrimental, particularly, in the center-city parts of the urban areas (ref. 20). Unlike urban O_3 , the relative effects of VOC and NO_x controls on regional O_3 have not been well studied yet. There is suggestive evidence that control of NO_x in urban and rural areas would reduce regional O_3 but more complete studies are necessary before such an effect can be ascertained and quantified.

The final comments pertain to the modeling tools currently available. From the discussion thus far, it can be seen that the O_3 problem is too complex for the control agencies to design ozone control strategies based on "common sense" or on empirical evidence alone. Notwithstanding their imperfections and limitations, state-of-the-art physicochemical O_3 air quality models are the superior tool, indispensable in O_3 air quality management practices. For the urban O_3 problem case, there are now available simple, EKMA-type models (refs. 26,27) as well as more sophisticated--but costlier--Eulerian models (ref. 28), that can be used by the scientific community within the air pollution control agencies in the US. The EKMA models are acceptable for small, isolated urban areas with mild O_3 problems whereas use of the grid models is generally preferable in the cases of severe O_3 problems in urban areas within densely populated regions. In either case, the input information required by the models is available or can be measured, except for future boundary conditions. The requisite data on this latter input can be estimated only through use of predictive regional models and this is one reason why regional O_3 models have been developed. Regional models are now available in the US and Europe but,

unlike the urban scale models, their use is still quite limited. Thus, in the US, for example, EPA's ROM has not been fully evaluated yet, the probabilistic nature of its predictions complicates the use of the model for O₃ strategy development, and lastly but most importantly, its complexity and cost of application make the model accessible to only a few. The model is extremely demanding in quantity and quality of input and in ADP resources. For example, the requisite emission inventory input should include biogenic emissions data, and the strong sensitivity of model predictions to the NO_x emission factor makes it crucial that the NO_x emission inventory input be extremely accurate. Finally, there are imperfections in the chemistry and dispersion components of the model. For example, the model's chemical mechanism does not include aerosol-related steps and, therefore, sink processes such as loss of H₂O₂ and of NO_x-derived products on liquid/solid particles, are ignored, and, as a result, model predictions for H₂O₂ and some NO_x species may be erroneously high. While it may take several years to remove all of these imperfections, it is expected that in a year or so the current version of USEPA's ROM will be fully evaluated and ready to be used for development of ozone control strategies.

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